

## Origin and fluxes of dissolved sulphate in the Himalayan system : evaluation and implication

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Continental weathering budget requires careful analysis of dissolved sulphate origin as sulphide oxidation may represent a source of acid auxiliary to CO<sub>2</sub> [1]. In Himalayan rivers the concentration of sulphate is about 10 meq% that of HCO<sub>3</sub><sup>-</sup> and previous studies underline the importance of sulphide oxidation processes [2-3]. We present isotopic compositions of dissolved sulphates from various Himalayan basins in Nepal, India and Bangladesh. More detailed data come from the Central Nepal Narayani basin. Isotopic data reveal that δ<sup>34</sup>S are low between +6 and -15‰ and δ<sup>18</sup>O are between +3 and -10‰. In addition there is a clear correlation between oxygen isotopic compositions of the river water and those of dissolved sulphate. These data confirm that the large majority of dissolved sulphate is derived from sulphide oxidation reactions occurring during erosion. Such reactions appear to be widespread over the Himalayan basin and can be sometime observed in winter as efflorescence of gypsum at the surface of sulphide bearing rocks such as black shale.

The analysis of rivers draining different lithologies in Nepal reveal that while sulphide oxidation is the prevalent source of sulphate it's intensity is highly variable. Assuming that all dissolved sulphate derive from pyrite, chemical erosion of pyrite vary between 10 and 60 t/yr/km<sup>2</sup> in High Himalayan gneisses and in Tethyan Sedimentary Series respectively. Among primary Himalayan basins, the Narayani Gandak and the Siang-Tsangpo rivers are the major sources of dissolved sulphate. Ganga and Brahmaputra in Bangladesh have isotopic compositions comparable to their Himalayan tributaries. The fluxes recorded in Bangladesh suggest that anthropogenic input to the river flux is not dominant in spite of the dense agricultural and industrial activity in the floodplain.

Overall sulphide oxidation processes represent a source of acid for weathering reactions during Himalayan erosion. It's association with Tethyan sediments suggest that sulphuric acid will mostly dissolve carbonates acting on the long term as a net source of CO<sub>2</sub> to the atmosphere.

[1] Calmels et al. (2007) *Geology* **35** 1003-6 [2]A. Galy & France-Lanord (1999) *Chem. Geol.* **159**, 31-60. [3] Karim & Veizer (2000) *Chem. Geol.* **170**, 153-177.

## Particle size dependence on arsenic concentration and bioaccessibility in mine tailings from the Empire Mine, CA

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As a result of the historic mining of gold and silver in California, an estimated 47,000 abandoned mine sites exist statewide. Since the halt of mining operations, communities have developed near and around abandoned mine wastes at these locations. This is of great concern because the waste material left over from the processing and extraction of precious metals contains associated toxic metal(loid)s, including arsenic (As), which can be mobilized into surrounding communities, contaminate surface and rain water, and be incidentally ingested by humans. The Empire Mine in the Sierra Nevada mountain range contains areas of varying As contamination. Preliminary studies have discovered a range of As bioaccessibility relative to initial As concentration, indicating that trends in arsenic concentration and bioaccessibility may be a function of particle size, correlations between arsenic and iron, and/or arsenic speciation.

Bulk samples were collected from six different locations at the Empire Mine and separated by particle size through a dry sieving method to generate eleven distinct size fractions. BET surface area analysis and EXAFS spectroscopy was conducted to determine the reactive surface area and arsenic speciation of selected size fractions. Acid digestion and ICP-AES analysis were then conducted to determine the initial concentrations of As and a host of other metals present in each size fraction. Finally, the bioaccessibility of As as a function of particle size was examined by conducting a physiologically based extraction test (PBET) utilizing a simulated gastric fluid (SGF).

The majority of samples collected displayed an inverse relationship between particle size and total arsenic concentration; these same samples also typically showed a strong As-Fe correlation. Conversely, samples without an inverse size-concentration relationship tend to have both low total As concentrations and poor As-Fe correlations. In addition, As concentrations were found to increase by over an order of magnitude as size decreases. In relation to the bioaccessibility tests, 7-10% As was consistently released into the simulated gastric fluid media, suggesting that bioaccessibility is linked with the initial As concentration of the sample. However, when the percent As released is normalized for surface area in comparison to particle size, a decreasing trend in As as particle size decreased resulted. This indicates surface area also plays a significant role in As bioaccessibility. Furthermore, bulk (unsieved) samples that undergo PBET can underestimate bioaccessibility in fine-grained size fractions, also supporting the possible effect that surface area and particle size play in the degree of arsenic bioaccessibility in mine wastes. These results demonstrate that particle size is an important criterion in determining arsenic transport, concentration, exposure, and future remediation efforts.